# Using New Fission Data with the Multi-Detector Analysis System for Spent Nuclear Fuel

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New experiments using an array of high purity germanium detectors and fast liquid scintillation detectors has been performed to observe the radiation emitted from the induced fission of $^{235}U$ with a beam of thermal neutrons. The experiment was performed at the Argonne National Laboratory Intense Pulsed Neutron Source. Preliminary observations of the data are presented. A nondestructive analysis system for the characterization of DOE spent nuclear fuel based on these new data is presented.

## 1 Introduction

Since the mid-1960s the experimental effort in nuclear structure and spectroscopy has taken a new direction that was opened with the development of the solid state radiation detector. The high-resolution gamma-ray spectra measured by these devices were seen to be the equivalent to the optical spectra in atomic physics. Although these solid state detectors brought new information and much higher energy resolution to the spectroscopy effort, correlation of different and multiple radiation emission using arrays of these detectors was slow in coming.

It was not until the mid- to late-1980s that large numbers of solid state detectors were used in arrays that could gather multiple types of time correlated radiation data. The high cost and complexity of these arrays limit the number that can be constructed and maintained world-wide. Today there are only three very large arrays (Gammasphere, Euroball, and Eurogam) and about a dozen smaller arrays[1]. These arrays of detectors have been the used exclusively in accelerator based nuclear reaction studies, except for a few exceptions in the past dozen years. An important set of experiments have been performed by this collaboration using these arrays for fission studies, which have resulted in discovering new information on the fission process.

## 2 Early Work

In the mid-1980s the first experiments [2-4] to study fission fragments using an array of high-purity germanium (HPGe) detectors were conducted at ANL. The

experiments used a $^{252}$Cf source and the Argonne-Notre Dame array of Compton suppressed HPGe detectors. Between 1990 and 1993 several other experiments using the Compact Array of 20 HPGe detectors at ORNL, the initial Gammasphere array at LBNL, and a small array of HPGe and x-rays detectors at INEEL were performed using sources of $^{252}$Cf and $^{242}$Pu. Unlike the initial experiment at ANL, these later experiments were run for long periods of time and from 100-200 gigabytes of computer data in list mode were collected. These data sets have been and continue to be rich sources of new information [5] on the fission process and the nuclear structure of the isotopes produced promptly in fission. This work resulted in the discovery of new gamma rays, isotopes, decays modes, neutron and gamma-ray multiplicity numbers, and other fundamental nuclear physics phenomena. The application of these results to arms control issues lead to the development of the Gamma-Neutron Assay Technique (GNAT) [6-8] to determine the isotopic ratios of fissile isotopes in nuclear weapons dismantlement. These fission studies and their new results are the basis of a non-destructive analysis (NDA) system under development to measure fissile mass, fissile isotopic ratios, radiation source term, and specific fission product isotopes. This system has applications for the Department of Energy Spent Nuclear Fuel (SNF) Program, Advanced Mixed Waste Focus Area, TRU Waste Program, Safeguards and Security Office, and Nonproliferation and National Security Office.

## 3 Current Work

In 1998 the Argonne-Notre Dame array of Compton suppressed HPGe detectors was moved to the ANL Intense Pulsed Neutron Source and installed to collect data on the induced fission of $^{235}$U with a beam of thermal neutrons. The electronics, computers, and software are from INEEL with IPNS providing laboratory space and a position on an IPNS beamline. The preliminary appraisal of the data collected in this experiment will be presented and how it will be used in the Multi-Detector Analysis System (MDAS) for NDA of DOE SNF.

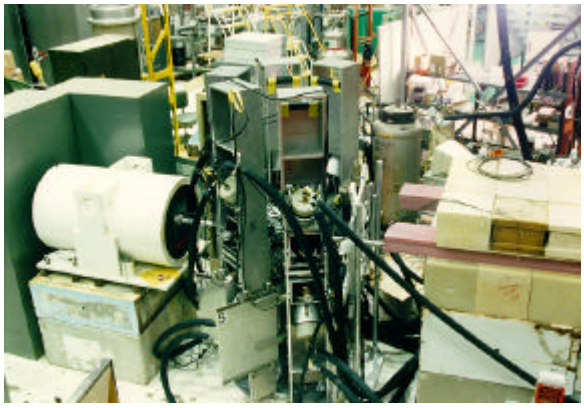

**Figure 1.** Scintillator and HPGe detector Array

## 4 Present Results

Figure 1 shows the experimental setup at the ANL/IPNS facility. The first experiment to use this new

setup called for placing a target of $^{235}U$ in the thermal neutron beam provided by IPNS and collecting data on the resulting induced fission events. This measurement was started in June 1998 and was allowed to run for a period of several weeks of beam time to obtain in excess of $10^9$ events. The data-taking phase for $^{235}U$ measurement ended in December 1998. The raw event data are stored on a computer medium, and will be analyzed to identify decay modes, gamma-ray signatures, nuclear structure, radiation multiplicities, and other relevant parameters involved with fundamental nuclear physics research.

**Table 1.** Fission Pairs with Gamma Rays Observed in Spectra but Partner is Unknown

| Observed Fission Isotope | Unknown Partner for 2N | Unknown Partner for 3N | Unknown Partner for 4N |
|---|---|---|---|
| $^{138}Ba$ (1435 keV) | $^{95}Kr$ | $^{94}Kr$ | $^{93}Kr$ |
| $^{140}Ba$ (602 keV) | $^{93}Kr$ | | $^{91}Kr$ |
| $^{140}Ce$ (1596 keV) | | $^{92}Se$ | |
| $^{142}Ce$ (641 keV) | | $^{90}Se$ | |
| $^{96}Zr$ (1750 keV) | | $^{137}Te$ | |
| $^{88}Sr$ | | $^{144}Xe$ | |

The plan is to follow the $^{235}U$ measurement with additional measurements involving other fissile isotopes important to the SNF effort. In addition to the continued data collection effort, analysis of the $^{235}U$ target data and the earlier spontaneous fission data will continue. The fact that new decay modes have been observed in the spontaneous fission raises the question, "Do these decay modes exist in induced fission?' This requires a comparison between the data sets.

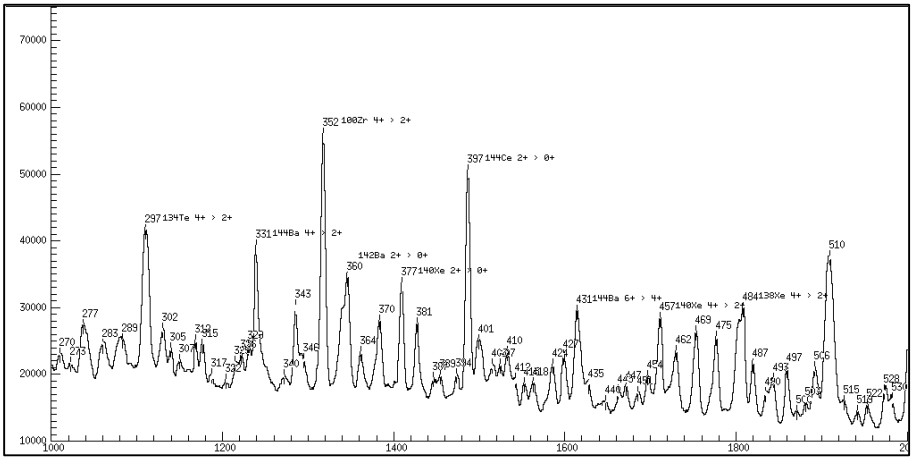


**Figure 2.**

An initial cursory examination of the total coincidence spectrum for one of the ten HPGe detectors has been made to check the quality of the data and plan the

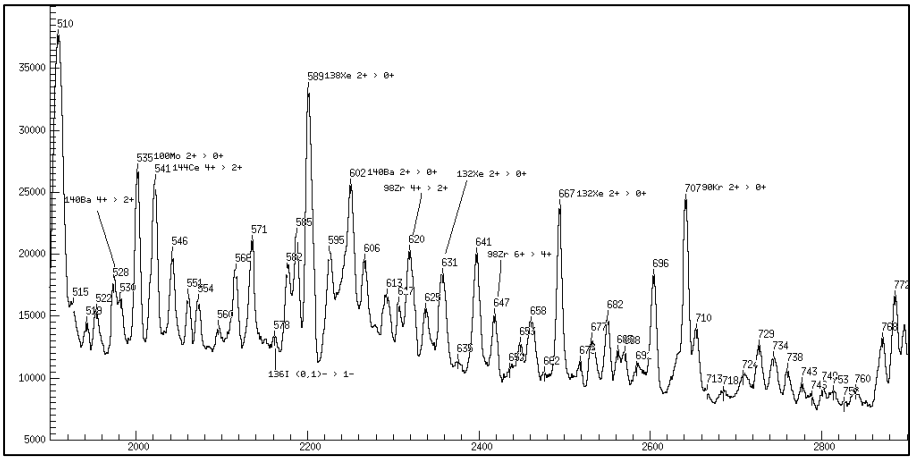


**Figure 3.**

analysis process. As an example of the new information that can be obtained from such a data set Table 1 show pairs of fission fragments for the cases where a

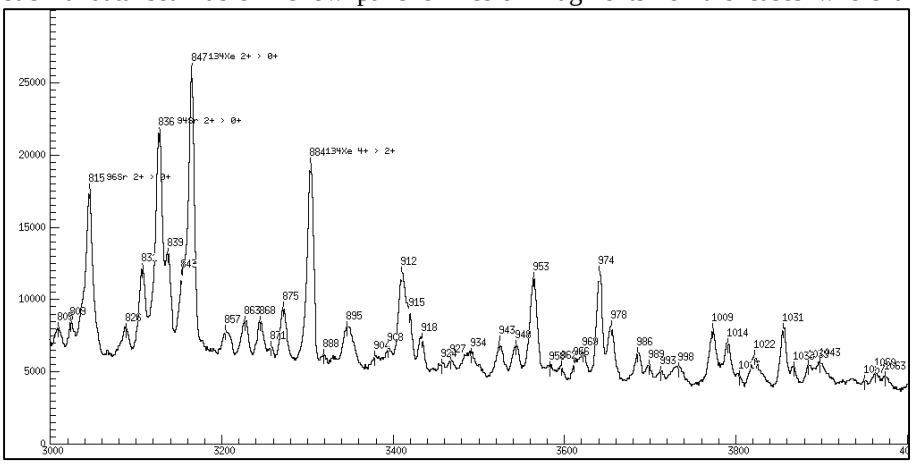


**Figure 4.**

transition in one of fragments has been observed in the total coincidence spectrum. The complimentary fragments must also be present but they have so far never been observed. Analysis of the present data should provide the first level diagrams for these nuclei.

Figures 2-5 show the first part of a spectrum that we examined. The numbers above the peaks are energies in keV, and some peaks have been identified with a

particular isotopes and its placement in the level scheme. The x-axis of the plot is in channel number and the y-axis is intensity in counts. These regions are presented to show the high quality of the data and support the premise that much new information can be revealed in the analysis.

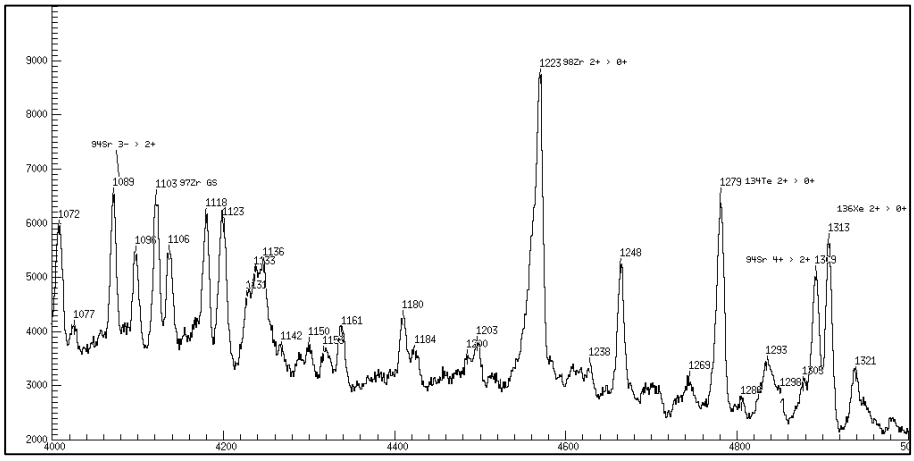


**Figure 5.**

## 5 Applications and Plans

The study of the fission process with the new multi-detector arrays, faster electronics and detectors, and the new generation of computers have already yielded stunning results on the fundamental understanding of the fission process. New decay nodes show a degree of freedom in the nuclear system that was not previously known. The direct isotope yields previously measured required the use of fast separation methods that introduced a bias in the data. The use of the arrays of detectors and the identification of the promptly produced isotopes removes this bias. The neutron multiplicity is obtained from the identification of the fission partners seen by the coincidence system. No ambiguity exists in this measurement. For the fundamental understanding of the fission process, these results have produced new information that is allowing specific channels and processes to be observed. Average characteristics values are being replaced with specific values in models and reported results.

The results of this experimental effort have direct application to new characterization and verification methods that have been and are being developed at INEEL. These NDA methods support DOE goals in environmental management and remediation in the areas of advanced mixed waste, transuranic (TRU) waste, SNF, safeguards, and arms control and national security. Specific characterization needs for moving SNF from wet to dry storage and final transportation and

acceptance of the DOE SNF at the repository will be fulfilled by the applications of results obtained in this research.

The next measurement planned is to collect induced fission data for $^{239}$Pu. Currently we have located a target of ~96% pure $^{239}$Pu which can be encapsulated for use at IPNS. In 1999, we will continue with these experimental efforts for as long as we have access to the detectors and the neutron beam at IPNS. The loan of the detector array will most likely end in 2000. We plan to search for other available material if funding is received.

## 6 MDAS

The Spent Nuclear Fuel (SNF) Non-Destructive Analysis (NDA) program at INEEL is developing a system to characterize SNF for fissile mass, radiation source term, and fissile isotopic content. MDAS is a new approach to the non-destructive analysis of fissile and other radioactive material. It uses the fundamental physics of fission and decay processes, very fast coincidence methods, electronics and computers available only in the past few years, and specially designed high-rate detectors. The time-correlated nature of signals from the array of detectors, of multiple types, is used to reduce the underlying spectroscopic backgrounds by orders of magnitude over previous methods. MDAS uses these methods to determine the ratios of fissile isotopes, and the total quantity of fissile material in the item, using an external source of neutrons. For MDAS the effects of extended source size, radiation attenuation, high backgrounds, and non-fissile radioactive material present will compensated for in the analysis software. The quantification of fissile material is determined from the gross counting of neutron coincidences instead of the identification of specific gamma-rays selected from a global spectrum. In addition to fissile quantity, MDAS determines radioactive source term, selected nuclide inventory, and uranium enrichment. Using highly-energetic radiation, MDAS will characterizes items such as spent fuel and transuranic waste without special calibration standards or a priori knowledge.

### *6.1 Background*

The fundamental difference between conventional radiation detection and measurement methods and the methods used by MDAS lie in how the physical process emitting the radiation is understood and the signals generated by the detectors are collected, processed, and stored. Traditional spectroscopy and radiation detection measure radiation as a "field" or a continuous emission of energy. In reality, each time a nucleus fission’s or emits radiation it is a single discrete "event" in time with individual quanta or particles of radiation. MDAS uses an event-by-event method to collect the data from the detectors, whereas traditional; spectroscopy accumulates an integrated signal. Treating radiation as a field came about because most radiation measuring equipment and detectors were

"slow" with respect to the radiation emission process. In the past twenty years, radiation detectors and their support electronics have been developed that can respond in the time scales of hundreds of picoseconds ($10^{-12}$ seconds). This means that such detectors can respond to individual elements of the emitted radiation even if the emission rate is in the giga-hertz range ($10^9$ Hz). For comparison, this means that the decay of a source of $^{137}$Cs of one R/hr at 10 cm could be monitored on an individual atom-by-atom basis.

The scientific expertise that is fundamental to MDAS has been developed and validated by the experimental nuclear physics community for many years. Innovations in computer equipment (reduced cost, reduced size, increase speed and power, networking, fiber-optic communications, and others) allow things to be done now in real-time or within hours of a measurement that could not be done just a few years ago. The power of computer programming languages, new programming development tools, and extensive use and availability of graphical systems and interfaces reduce the need for a specialist to make NDA measurements or analyze the data. The basis and techniques of the data acquisition and analysis was developed by the physics community to satisfy the requirements of more complex and costly experiments. In addition to new equipment, new methods for NDA use are now available. The methods used in MDAS are coincidence spectroscopy with large detector arrays [1,9,10], coincidence gating [11-13], list-mode data acquisition [14], fast liquid scintillators [15-18], and pulse-shape discrimination [19-21]. These combined with the new understanding of the radiation process will be addressed below.

### *6.2 Radiation Sources*

The approach used by MDAS incorporates a new understanding of the physics of radiation sources. The new understanding emerges from the correlation of the prompt radiation (gamma rays and neutrons) that occur in the fission process and the multiple gamma-rays that are produced by the prompt de-excitation of the isotopes produced in fission. For the case of fission, "prompt" means a time period of approximately $10^{-22}$ seconds. The fission products are: 1) two fragments, one light mass and one heavy mass; 2) zero to 10 evaporation neutrons; and 3) several de-excitation gamma rays. The heavy-mass fragments with masses from approximately 130 mass units (A = 130) to about 145 mass units (A = 145) can have yields of 2-5% each of the total mass yield, independent of the fissioning nuclei. Except for very rare cases (approximately $1{:}10^8$), fission is a binary process with the energetic (e.g., easily observable) radiation coming from the two fission fragments. The gamma rays observed as being part of the fission process are due to the de-excitation of the fragments. In previous studies, the conventional wisdom has been that the energies of gamma rays from either of the two prompt fragments lie predominately between 300 to 400 keV. This assumption was a result of the methods used in studying the fission fragments.

Until the mid- to late-1980s, fission fragments were never studied directly because of instrumental limitations that existed. Studies [22-35] undertaken by the INEEL nuclear physics staff in collaboration with partners from other DOE laboratories and universities have shown that high-energy gamma-rays come from fission fragments near the doubly closed shell at Z=50 and N=82. These gamma rays can be used by a coincidence system to identify the fissioning element. Additional analysis will identify the specific isotope undergoing fission.

### *6.3 Example*

The even-even isotopes of $^{132,134}$Sn and $^{134,136}$Te have not only a very high yield, but also multiple gamma-ray transitions with energies of 2-4 MeV. An example is shown in Figure 6 for $^{134}$Te isotope, which, in the case of $^{240}$Pu, makes up about 5% of the fission fragment yield. We see two very high-energy transitions of 2.322 and 2.865 MeV feeding the third excited state at 1.693 MeV. In the case of fission, the neutrons and gamma rays are all in coincidence, observing these high-energy gamma rays from one fragment makes it possible to observe the gamma rays in the partner fragment. By observing the gamma rays from both fragments, the Z of the fissioning elements is uniquely identified. By observing the ratio of different light-mass fragment produced, the masses (A) of the specific fission isotopes and their ratios can be determined.

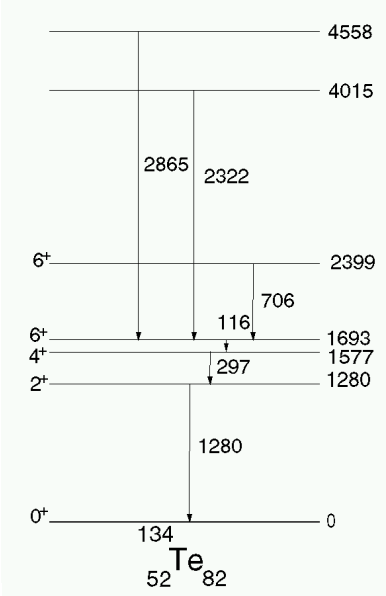


**Figure 6.** Levels in $^{134}$Te showing new transitions

Figure 7 shows the currently accepted levels in $^{134}$Te[36]. The two high-energy gamma rays at 2.322 and 2.865 MeV have not been reported previously.

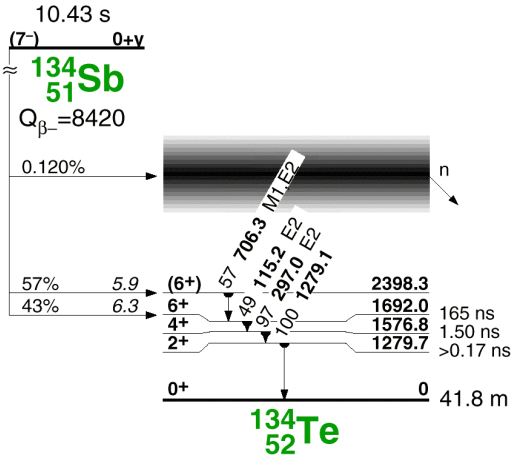


**Figure 7.** Levels of $^{134}$Te known from beta-decay [36]

*6.4 Conservation Rules*

The important point of the method is that for binary fission the two fragments account for the total Z of the original fissioning element. This results in a charge (Z) conservation between the two fragments and the initial fissile isotope.

$Z_F = Z_H + Z_L$ Z Conservation

The two fragment isotopes and the neutrons emitted account for the mass number of the nucleus that fissioned. This leads to the mass number conservation that is seen in a distribution of the masses of the fragments.

$A_F = A_H + A_L + xn$ A Conservation

Table 2. Selecting the gamma rays of a fragment identifies the fission partner in a coincidence gate. The number of protons in the two fragments must sum to the number of protons in the fissioning element. For the fission isotopes listed, the isotopes that are partners for the cases of zero to five emitted neutrons are listed for the $^{134}$Te isotope.

| Fissioning Isotope | $T_{½}$(SF) yr | Light-mass Fragment Paired with $^{134}$Te | | | | | |
|---|---|---|---|---|---|---|---|
| | | N=0 | N=1 | N=2 | N=3 | N=4 | N=5 |
| $^{235}$U | $3.5X10^{17}$ | $^{101}$Zr | $^{100}$Zr | $^{99}$Zr | $^{98}$Zr | $^{97}$Zr | $^{96}$Zr |
| $^{238}$U | $8.19X10^{15}$ | $^{104}$Zr | $^{103}$Zr | $^{102}$Zr | $^{101}$Zr | $^{100}$Zr | $^{99}$Zr |
| $^{239}$Pu | $5.5X10^{15}$ | $^{105}$Mo | $^{104}$Mo | $^{103}$Mo | $^{102}$Mo | $^{101}$Mo | $^{100}$Mo |
| $^{240}$Pu | $1.34X10^{11}$ | $^{106}$Mo | $^{105}$Mo | $^{104}$Mo | $^{103}$Mo | $^{102}$Mo | $^{101}$Mo |
| $^{241}$Am | $1.15X10^{14}$ | $^{107}$Tc | $^{106}$Tc | $^{105}$Tc | $^{104}$Tc | $^{103}$Tc | $^{102}$Tc |
| $^{242}$Pu | $6.75X10^{10}$ | $^{108}$Mo | $^{107}$Mo | $^{106}$Mo | $^{105}$Mo | $^{104}$Mo | $^{103}$Mo |
| $^{252}$Cf | 82.8 | $^{118}$Pd | $^{117}$Pd | $^{116}$Pd | $^{115}$Pd | $^{114}$Pd | $^{113}$Pd |

Table 2 lists the light-mass fragments that are associated with the heavy-mass fragment $^{134}$Te for the fission isotopes listed in the first column. The fission half-life, in years, is included in the table as well as the specific isotopes for the cases of zero to five emitted neutrons.

The same studies [22-35] referenced previously have shown that a multiple detector array can be used to detect pairs of these fast neutrons in coincidence and determine the fissile mass present to within 50%.

## 7 MDAS Description

A prototype multi-detector analysis system is being developed that will have 68 detectors; 20 high-purity germanium (HPGe) for gamma-ray detection and 48 liquid scintillaor detectors for neutron detection. The liquid scintillators use pulse-shape-discrimination. This system has several advantages: fast coincidence methods, list-mode data, gamma-ray coincidence, neutron coincidence, pulse-shape discrimination, detector arrays, and data acquisition and analysis. It is important to note that MDAS integrates all the individual advantages and performs the measurement to obtain the gamma-ray and neutron data in a single operation.

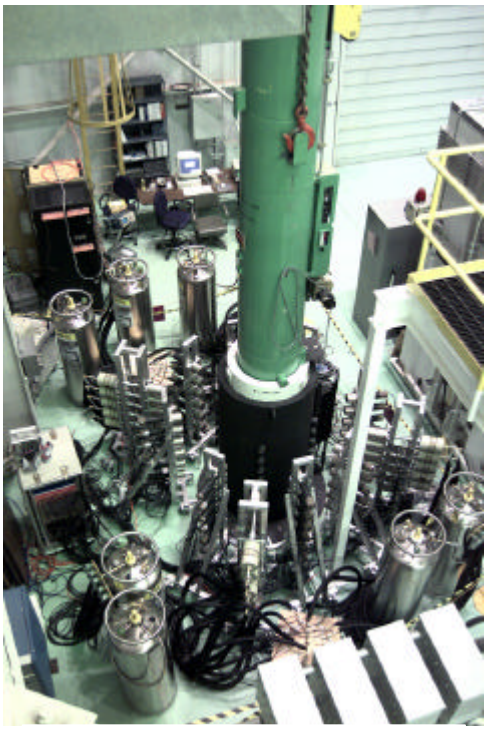

**Figure 9.** MDAS prototype.

## 8 Conclusions

The prototype system is the test-bed to refine the basic methods and characteristics, such that specific information can be obtained for the spent nuclear fuel and transuranic waste programs of DOE. The basic physics has been proven, and, in some special cases (arms control verification), particular systems have been developed as demonstrations. The successful prototype will also serve as the basis for an operational system, part of which involves the customization of methods and hardware for specific tasks and results.

Summarizing the important characteristics:

- Prototype uses 68 detectors
- Neutron coincidences are used to determine fissile mass
- Neutron detectors are organic liquid scintillators, very fast response
- PSD allows selection of neutron coincidence events
- Gamma-ray detectors are specially designed HPGe

- Only coincidence events are acquired as data
- Short coincidence time window reduces random background
- Data are acquired, analyzed, and stored as list-mode format
- List-mode format is easily archived, retrieved, and re-analyzed
- Software gating selects only data of interest for analysis
- Distributed computing approach separates data acquisition and analysis

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